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Author(s):

D. M. Gilliam, J. A. Grundl, and G. E. Hansen

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D. M. Gilliam
J. A. Grundl

National Bureau of Standards

Gaithersburg, Maryland 20760

G. E. Hansen

Los Alamos Scientific Laboratory of the University of California

Los Alamos, New Mexico 87545

ABSTRACT

Measured average reaction cross sections for the Big Ten central flux spectrum are given together with calculated values based on the U.S. Evaluated Nuclear Data File ENDF/B-IV. Central reactivity coefficients for 233U, 235U, 239Pu, 6Li, and 10B are given to check consistency of bias between measured and calculated reaction cross sections for these isotopes. Spectral indexes for the Los Alamos 233U, 235U, and 239Pu metal critical assemblies are updated, utilizing the Big Ten measurements and interassembly calibrations, and their implications for inelastic scattering are reiterated.

REACTION RATE MEASUREMENTS IN BIG TEN

As part of the Interlaboratory Reaction Rate (ILRR) Program¹ to measure reaction rates in a variety of neutron fields, standard Big Ten sample irradiations were made for the participating laboratories: Rockwell International for helium production in ⁶Li and ¹⁰B; National Bureau of Standards (NBS) for fissions in ²³³U, ²³⁵U, ²³⁸U, ²³⁷Np, and ²³⁹Pu; Argonne National Laboratory, Atlantic Richfield Hanford Company, Hanford Engineering Development Laboratory, and Idaho Nuclear Engineering Laboratory

for the γ -ray measurements. Additionally, there were standard sample irradiations for the radiochemistry group of the Los Alamos Scientific Laboratory (LASL) for activation measurements. Fluence monitoring was principally by the centrally located NBS fission chamber and, secondarily, by externally located fission chambers and centrally located activation foils.

Table I lists the results of these measurements and the results of calculations based on the U.S. Evaluated Nuclear Data File, ENDF/B-IV. The experimental data, originally expressed as reactions per second per atom, have been scaled to make the ILRR measured ^{235}U fission rate numerically equal to 1375 which corresponds to the calculated integral fission cross section in millibarns. Uncertainties in measured integral cross section ratios are then simply obtainable by compounding the uncertainties listed for the individual reactions. The agreement between reaction rates obtained by the ILRR laboratories and by the LASL is good save for $^{115}\text{In}(n,n')$ ^{115}mIn and $^{27}\text{Al}(n,\alpha)$ ^{24}Na .

In the case of the indium reaction, part of the disagreement is caused by the choices of different γ branching ratios: for the 336 keV gamma, ILRR used an intensity of 45.9% and LASL used 47.3% both of which differ from the value 49.5% used on the ENDF/B-IV cross section evaluation. If one redetermines the LASL and ENDF/B-IV integral cross sections with the ILRR gamma intensity, the $^{115}In(n,n')$ ^{115m}In values are respectively 35.0 and 36.1 mb. In the case of the aluminum (n,α) reaction, there is a 13% difference between ILRR and LASL values, a difference essentially equal to that between values obtained by CEN/SCK and ULhC for the $\Sigma\Sigma$ neutron field.⁶ One concludes that errors in absolute γ-detection efficiencies for the relatively high energy 24Na gammas prevent meaningful comparisons of aluminum (n,α) integral cross sections measured for different neutron fields by different laboratories unless error correlation is introduced by intercalibration of the detection system.

INTEGRAL CROSS SECTION CALCULATIONS FOR BIG TEN

The central flux spectrum was calculated by a multigroup neutron transport code using an S_4 quadrature, ENDF/B-IV material cross sections processed into 0.125 lethargy width groups, and a Maxwellian fission neutron spectrum with a temperature of 1.336 MeV. As with other simple assemblies, the calculated flux spectrum above a few MeV has essentially the same energy dependence as the input fission neutron spectrum regardless of choice of material cross sections. Although one should have used an input spectrum very close to the thermal neutron induced fission neutron spectrum, χ_{25} , of 235 U, the significant difference is presumably in the high-energy tails.

Big Ten Dosimetry

TABLE I. INTEGRAL CROSS SECTIONS IN THE BIG-10 CENTRAL FLUX SPECTRUM

Reaction, R	-	Measured (mb)					Calculated (mb)	<u>Ref</u> s.
		ILR	R	LASL			ENDF/BIV	
⁵⁹ Co(n,Y) ⁶⁰ Co	13.0	±	0.2	12.8	±	0.5	12.6	1,2
⁵⁸ Fa(n,Y) ⁵⁹ Fe	4,21	±	0.11	4.00	±	0.16	3.09	1,2
55 _{Mn(n,Y)} 56 _{Mn}				7.4	±	0.2		2
63 _{Cu(n,Y)} 64 _{Cu}	22.5	±	1.3	23.8	±	1.4	24.8	1,2
197 Au(n, Y) 198 Au	230	±	2	235	±	4	220	1,2
238 _{U(n,Y)} 239 _U	151	±	4	1.17	±	3	148	1,2
10 _B (n, He)	1380	±	4			_	1217	3
45 Sa(n, Y) 46 Sc	18.2	±	0.3	17.5	±	0.5	15.9	1,2
$^{113}In(n,\gamma)^{114}In$			•	583	±	35		2
115 _{In(n,Y)} 116 _{In}				200	±	6	233	2
6Li(n, He)	966	±	4				953	3
233 _{U(n,f)}	2214	±	50				2091	4
235 _{U(n,f)}	≡ 1375	±	15	1391	±	28	1375	5
239 _{Pu(n,f)}	1647	±	18		_		1613	5
237 _{Np(n,f)}	436	±	8				460	5
115 _{In(n,n')} 115 _m _{In}	37.2	±	0.7	34.0	±	1.7	33.5	1,2
236 _{U(n,f)}	51.3	±	0.7	52.1	±	2.6	52.4	5,2
47 Ti(n,p) 47 Sc	2.96	±	0.11	2.85	±	0.11	3.55	1,2
58 _{Ni(n,p)} 58 _{Co}	16.9	±	0.3	16.2	±	0.5	16.2	1,2
54 Fe(n,p) 54 Mn	12.4	ŕ		11.9	±	0.4	12.1	1,2
46 _{Ti(n,p)} 46 _{Sc}	1.79	±		1.77	±	0.05	1.56	1,2
²⁷ A1(n,p) ²⁷ Mg		_	0.03	0.545	±	0.01		
56 _{Fe(n,p)} 56 _{Mn}				0.128	±	0.009	_	2
238 _{U(n,2n)} 237 _U						_		2
27 Al(n, a) 24 Na	0.405		0 000	0.204	±	U.008		2
48 Ti(n,p) 48 Sc	0.107		0.003	0.095	±	0.00		1,2
	0.049	±	0.001	0.048	±	0.00	0.032	1,2
¹⁹⁷ Au(n,2n) ¹⁹⁶ Au				0.49	±	0.05		2

Integral reaction cross sections were determined from the calculated central flux spectrum and multigroup cross sections derived from ENDF/B-IV. For radiative capture in ²³⁸U, the shielded multigroup cross sections processed for the transport calculations were used. We believe the calculated integral cross sections for the non-threshold reactions properly reflect the nuclear data taken from ENDF/B-IV. For reactions with thresholds progressively higher than that of ²³⁸U(:,f), the calculated integral cross sections depend progressively more strongly on the

fission neutron spectrum used in the flux calculation. Here direct comparison between calculations and measurements of the best known high-threshold reactions should expose the flaws in the input spectrum even as similar calculations and measurements for χ_{25} should expose the flaws in the best present description of that basic spectrum.

We wish to confine our attention in the remainder of this section to the non-threshold reactions of Au, ^{10}B , ^{6}Li , ^{233}U , ^{235}U , and ^{239}Pu where there are supportive data to those of Table I. As an example, the absolute fission ratio measurements by Amundson et al. for the Flattop-25 spectrum and by Linenberger and Lowry for the Topsy spectrum, together with relative fission ratio measurements connecting these to Big Ten, give, for Big Ten, the integral cross section ratios $\sigma_f(^{233}\text{U})/\sigma_f(^{235}\text{U})$ = 1.564(±1.6%) and $\overline{\sigma}_f(^{239}\text{Pu})/\overline{\sigma}_f(^{235}\text{U})$ = 1.170(±1.6%); the corresponding ILRR values from Table I are 1.610(±2.5%) and 1.198 (±1.5%). We use a weighted average of these data in Table II which lists measured and calculated reaction cross section ratios and reactivity coefficient ratios. 10

The biases between measured and calculated fission cross section ratios for the fissile isotopes are essentially duplicated by the biases between measured and calculated reactivity coefficient ratios for Big Ten, and again by the biases between measured and calculated k-eigenvalue-ratios for the bare ²³³U, ²³⁵U, and ²³⁹Pu metal critical assemblies. For these latter, harder spectrum, assemblies, the biases between measured and calculated fission ratios and reactivity coefficients are very close to those for Big Ten. These spectrum insensitive results indicate biases in the ENDF/B-IV implications of fission ratios but, by themselves, reveal nothing more specific.

The two reasons for including ^6Li in Table II are 1) to show a helium production measurement result that is consistent with a reactivity coefficient measurement result and 2) to show that the calculations using $^6\text{Li}(n,\alpha)$ cross sections from ENDF/B-IV are in reasonable agreement with these results.

The observed helium production cross section of ¹⁰B exceeds the calculated value by a surprising margin. The observed reactivity coefficient confirms that the neutron absorption cross section for ¹⁰B is indeed larger than the calculated value although the two observations are only in qualitative agreement. These results perhaps suggest that the central flux spectrum of Big Ten is softer than the calculated spectrum but this is not confirmed by the gold reaction measurements which are only marginally different from calculations. That is, it is difficult to see how any readjustment of the calculated Big Ten spectrum could yield agreement between measured and calculated reaction cross sections for both ¹⁰B and ¹⁹⁷Au.

TABLE II. Comparisons of Reaction Rate and Reactivity Coefficient Ratios in the Center of Big Ten.

	Measured 0	alculated	Measured/Calculated
$\sigma_{\rm r}(^{233}{\rm U})/\sigma_{\rm r}(^{235}{\rm U})$ $\Delta k_{\rm o}(^{233}{\rm U})/\Delta k_{\rm o}(^{235}{\rm U})$	1.580(±1.95)	1.521	1.039 ± 0.019 ⁸
$\Delta k_o(^{233}U)/\Delta k_o(^{235}U)$	1.729(±1.0%)	1.677	1.031 ± 0.010
σ _e (²³⁹ Pu)/σ _e (²³⁵ U)	1.185(±1.7\$)	1.174	1.009 ± 0.017 ^b
σ _f (²³⁹ Pu)/σ _f (²³⁵ U) Δk _g (²³⁹ Pu)/Δk _g (²³⁷ U)	1.658(±0.8\$)	1.626	1.020 ± C.008
ση, He(⁶ Li)/σ _f (²³⁵ U) Δk _g (⁶ Li)/Δk _g (²³⁵ U)	0.703(±1.2%)	0.693	1.014 ±0.012
Δk ₀ (⁵ L1)/Δk ₀ (²³⁵ U)	-0.389(±2.6\$)	0.375	1.037 ± 0.027
on, He(10B) /o _f (235U)	1.003(±1.2 \$)	0.885	1.133 ±0.013
σn,He(10B)/σf(235U) $Δko(10B)/Δko(235U)$	-0.513(±0.6%)	-0.472	1.087 ± 0.007
$\sigma_{-1}(Au)/\sigma_{s}(^{235}U)$	0.1676(±1.2 %)	0.1600	1.048 ±0.013
$\sigma_{n,\gamma}(Au)/\sigma_{f}(^{235}U)$ $\Delta k_{o}(Au)/\Delta k_{o}(^{235}U)$	0.1229(±1.0%)		1.004 ± 0.010

The measured to calculated eigenvalue ratio k(Jezebel-23)/k(Godiva) is 1.044 \pm 0.002.

CENTRAL SPECTRAL INDEXES FOR BIG TEN AND RELATED BENCHMARK ASSEMBLIES

The fission ratios $\overline{\sigma}_f(^{238}\text{U})/\overline{\sigma}_f(^{235}\text{U})$ and $\overline{\sigma}_f(^{237}\text{Np})/\overline{\sigma}_f(^{235}\text{U})$ have long been used as spectral indexes related to neutron flux fractions above the ^{238}U and ^{237}Np fission thresholds. The absolute ratio measurements in Big Ten together with similar measurements in Flattop-25 and χ_{25} 8,11 permit an updating of the observed spectral indexes for the ^{233}U , ^{235}U , and ^{239}Pu metal critical assemblies and associated fission neutron spectra reported by Grundl and Hansen some twelve years ago. 12 Table III lists the present "best" ratio data along with calculated ratios based on ENDF/B-IV cross-sections and Maxwellian fission neutron spectra (T = 1.35 MeV for the uranium assemblies and T = 1.41 MeV for the plutonium assemblies).

b The measured to calculated eigenvalue ratio k(Jezebel-Pu)/k(Godiva) is 1.007 \pm 0.002.

The agreement or disagreement between an observed and calculated spectral index is markedly affected by the choice of the fission neutron spectrum used in the calculation. The calculated ratio of the spectral index for an assembly spectrum to the spectral index for the fission neutron source spectrum is insensitive to the choice of source spectrum and depends principally on the inelastic transfer cross-sections of the assembly material. Thus, from Table III, it is seen for example that the calculated flux fractions above the ²³⁸U fission threshold for Godiva and Big Ten are too high relative to that of the X₂₅ source spectrum, and hence that inelastic transfer cross sections of ²³⁵U and ²³⁸U implied by ENDF/B-IV are too small. ¹³

To the extent one believes that the spectrum calculations properly account for the subtle effects of energy-dependent neutron diffusion or leakage on the spectral indexes, these calculations and the observed spectral indexes permit inference of the material inelastic cross sections $\sigma_{28}^{\rm in}$ and $\sigma_{37}^{\rm in}$ of thin-shell transmission experiments fame. These cross

TABLE III. OBSERVED AND CALCULATED SPECTRAL INDEXES OF BIG TEN, THE 235U, 233U, and 239Pu METAL CRITICAL ASSEMBLIES, AND OF THE CORRESPONDING FISSION NEUTRON SPECTRA.

	σ _f (238υ)/σ _f (235υ)			$\overline{\sigma}_{\mathbf{f}}(237_{\mathrm{Np}})/\overline{\sigma}_{\mathbf{f}}(235_{\mathrm{U}})$			
	0bs	Calca	Calc Obs	Obs	Calc ^a	Calc Obs	
X ₂₅	0.254(±2.2%)	0.2429	0.96	1.091(±2.4%)	1.068	0.98	
Godiva	0.165(±1.1\$)	0.1687	1.02	0.837(±1.6%)	0.877	1.05	
Flattop-25	0.149(±1.0%)	0.1522	1.02	0.765(±1.5\$)	0.809	1.06	
Big Ten	0.0373(±1.0%)	0.0389	1.04	0.316(±1.5%)	0.339	1.07	
X ₂₃	0.259(±2.4%)	0.2429	0.94	1.098(±2.4%)	1.068	0.97	
Jezebel-23	0.213(±1.1%)	0.1903	0.89	0.977(+1.6%)	0.929	0.95	
Flattop-23	0.191(± 1.0%)	J. 1684	0.88	0.892(± 1.6%)	0.849	0.95	
Хцд	0.263(± 2.4 \$)	0.2525	0.96	1.103(± 2.4 \$)	1.082	0.98	
Jezebel-Pu	0.214(± 1.1%)	0.1923	0.90	0.962(±1.7\$)	0.972	0.97	
Flattop-Pu	0.180(± 1.0\$)	0.1642	0.91	u.842(± 1.6≴)	0.825	0.98	

The calculations used ENDF/BIV cross sections and Maxwellian fission neutron spectra characterized by the temperatures 1.35 MeV for uranium and 1.41 MeV for plutonium.

sections, corresponding roughly to the inelastic transfer cross sections from above to below the ^{238}U and ^{237}Np fission thresholds for fission neutrons, were evaluated in Ref. 12, and, with the aid of Table III, we update and extend that evaluation to include the data from Big Ten.

<u>x</u>	σ ir 28	^l (x)	$\sigma_{37}^{in}(\mathbf{x})$			
	ENDF/B-IV	From Table III	ENDF/B-IV	From Table III		
239 _{Pu}	1.20	0.80±0.17	0.60	0.57±0.18		
233 _U	1.36	1.01±0.15	0.73	0.60±0.15		
235 _U	1.10	1.44 ±0.15	0.54	0.85±0.11		
238 _U	1.99	2.24±0.09	0.96	1.09±0.06		

The inferred inelastic cross sections for the fissile isotopes agree well with the values of Ref. 12. In the above table, the isotopes are listed in decreasing order of their plateau fission cross sections and, historically one believed this would correspond to an increasing order of their inelastic scattering cross sections σ_{28}^{10} and σ_{37}^{11} . The spectral index data, for the simple critical assemblies considered in this report, confirm that belief much more consistently than do the evaluated data from ENDF/B-IV.

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In summary, we have noted some discrepancies between dosimetry measurements and calculations based on ENDF/B-IV such as for the gold and boron-10 reaction cross-sections in Big Ten and for the inelastic cross sections of 233 U, 235 U, 238 U, and 239 Pu for fission neutrons. We understand that the discrepancy between measured and calculated fission cross section ratios for 233 U, and 239 Pu in Big Ten essentially disappears if calculations are based on the new ENDF/B-V. The influence of ENDF/B-V on other discrepancies remains to be seen.

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